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New Correlated-Electron Ground State in YbAgGe

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Recently, an unusual correlated-electron ground state was observed in the new compound YbAgGe. The magnetic susceptibility, which was measured on a single-crystalline sample down to 1.8 K, is anisotropic with a Curie-Weiss like temperature dependence at high temperatures. For the crystalline average, the effective moment is ~4.1μ_B, which is ~10% below the value predicted by Hund's rules, and the Weiss temperature is -47 K, suggesting antiferromagnetic correlations are present. When the applied field is oriented along the c-axis of the tetragonal crystal structure, a broad maximum was observed around 3 K.

This behavior in the susceptibility may be a precursor of an unusual ground state that develops at lower temperatures. A maximum was observed in the specific heat at ~1 K with a very sharp peak just below the maximum at 0.62 K. The features in the specific heat could be interrupted as the formation of a heavy-Fermi liquid with a Sommerfeld coefficient of ~1.5J/mol K^2 followed by the development of long range order at lower temperatures. Even though no hysteresis or latent heat was observed, the abruptness of the anomaly at 0.62 K seems to imply some first order character maybe associated with this phase transition. Below the transition a significant residual electronic contribution seems to remain though this statement is very uncertain because it is based on data down to only half the transition temperature. If this transition is magnetic, then the properties of YbAgGe could result from a competition between long-range magnetic order and hybridization associate with localized spin fluctuations.

Finally, the electrical resistivity monotonically decreases with decreasing temperature in a fashion that is similar to other Yb-based heavy fermion compounds. The magnetoresistance was measured in fields up to 18 T, and at low temperatures, it is negative in contrast to the positive magnetoresistance observed for the nonmagnetic analog LuAgGe, implying that scattering from localized moments is important.

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Magnetization and de Haas-van Alphen Measurements to 50 T on UCd_{11}

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We have performed magnetization and de Haas-van Alphen (dHvA) measurements in pulsed fields to 50 T on the heavy fermion antiferromagnet UCd_{11}. The measurements were performed along the (100) axis of the cubic crystal. The electronic specific heat coefficient of 840 mJ/mol K^2 for UCd_{11} is the highest for any magnetically ordered uranium heavy fermion compound. At the lowest temperature measured (0.49 K) magnetic transitions at an applied magnetic field of B_{M1} = 6 T and B_{M2} = 16 T occur as seen in Figure 1. When the signal is plotted versus the inverse field, one observes oscillations in the signal with a frequency that is directly proportional to an extremal area of the Fermi surface; this is the dHvA effect. In the insets of Figure 1, there are plots of the fast Fourier transform (fft) of the signal versus inverse field for Region 1 ($B_{M1} < B < B_{M2}$) and Region 2 ($B > B_{M2}$). From these plots, we observe four frequencies in Region 1 that correspond to four of the ten frequencies observed in Region 2. Due to the overlap in the observed frequencies, we conclude that the magnetic transition at 16 T does not greatly affect the Fermi surface of UCd_{11}. The masses of the dHvA frequencies in Region 2 range from 2-17 m_0.

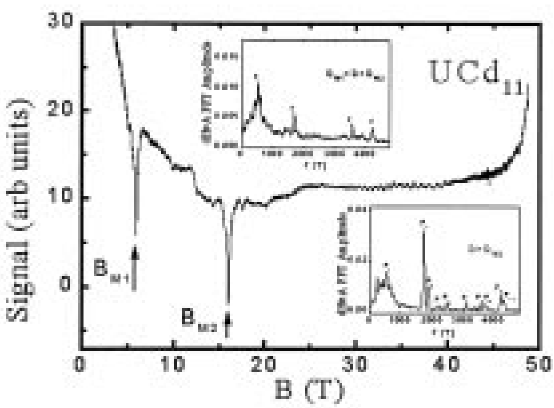

Figure 1. The measured signal versus applied pulsed magnetic field for UCd_{11} along the (100) axis at 0.49 K. The insets are fast Fourier transforms of the de Haas-van Alphen signal measured above and below the magnetic transition at B_{M2} = 16 T.

High Field Magnetoresistance of the Non-Fermi-Liquid Alloys $U_{1-x}Th_xPd_2Al_3$ and $U_{1-x}Y_xPd_2Al_3$

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We have performed magnetoresistance measurements at the NHMFL-LANL using a ^{3}He-^{4}He dilution refrigerator in the temperature range 20 mK ≤ T ≤ 2.5 K and a 20 T superconducting magnet. Measurements were made on two different kinds of uranium-based materials that exhibit non-Fermi-liquid behavior in their physical properties at low temperatures, $U_{1-x}Th_xPd_2Al_3$ and $U_{1-x}Y_xPd_2Al_3$. Previous measurements of R vs. H on the $U_{1-x}Th_xPd_2Al_3$ system for x = 0.4 and 0.6 revealed the existence of a small negative magnetoresistance for fields up to 18 T and a field dependence that is strongly suggestive of a contribution from single ion Kondo scattering. New measurements of R vs. T on the $U_{1-x}Th_xPd_2Al_3$ system confirm the negative magnetoresistance and yield a resistivity that has a linear temperature dependence at low temperatures for x = 0.6 and an approximately quadratic temperature dependence at low temperatures for x = 0.4 and x = 0.2. The x = 0.4 sample has an upturn at low temperatures, which is likely due to Kondo scattering. At low temperatures and in fields between 10 T and 14 T, the quadratic temperature dependence of the resistivity in the x = 0.4 and x = 0.2 samples is interrupted by striking features that appear to be due to magnetic correlations. The origin of these features has not yet been established. Measurements of R vs. T on a $U_{0.2}Y_{0.8}Pd_2Al_3$ sample exhibit a resistivity that has a linear temperature dependence at low temperatures. This sample also develops an interesting feature at low temperatures and in fields between 10 T and 14 T which, again, may be due to magnetic correlations, although this has not yet been substantiated.

Properties of Hexaboride Materials

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The rare earth and alkaline earth hexaborides form in a simple cubic structure and show a remarkable variety of interesting behavior. We have been pursuing research into three different hexaboride systems.

The first is EuB_6. This is a ferromagnetic semimetal, with Curie temperature T_C = 15 K. It undergoes a metal to semimetal transition at the magnetic ordering temperature. We have shown that the plasma frequency shifts by a factor of two at this transition.[1] De Haas-van Alphen measurements have found four frequencies that are consistent with band structure calculations, but these calculations cannot explain the large plasma frequency shift at 15 K. Doping experiments substituting La for Eu show strong polaronic effects. These are also evident in Raman scattering experiments.[2]

A second set of experiments is a doping study of SrB_6 with Sm. We have found that at the few percent level, Sm forms a Kondo singlet with T_K = 3 K and low temperature specific heat γ = 1050mJ/mole-SmK^2.[3] This provides information for how the insulating state in SmB_6 may be developing.

A third line of research is deHaas-van Alphen measurements in the system $La_{1-x}Ce_xB_6$. Much is known about the end points, but we for the first time have been able to track the evolution of the dense Kondo state out to x=.6, and have our further experiments will attempt to take this to higher x. A complete dilution study in a heavy Fermion system such as this has not been done before. Its importance lies in the possibility that we may be able to see fairly directly how the Ce-Ce interactions develop as x increases. These interactions have been difficult to access experimentally and are believed to be crucial to the physics of heavy Fermion materials.

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De Haas-van Alphen Measurements on $(La_{1-x}Ce_x)B_6$

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Following our measurements on CeB_6 we decided to try to follow the details of the onset of the heavy fermion state in La-Ce hexaboride alloys. Samples of 0 to 60% Ce substitutions for La in $(La_{1-x}Ce_x)B_6$ were made, and last summer and autumn we performed preliminary measurements in the 60 T pulsed field facility at the NHMFL in Los Alamos. The results for the 10% sample with the field applied along the [100] sample direction are shown in Figure 1. In Figure 2, the Fourier transform of the data in

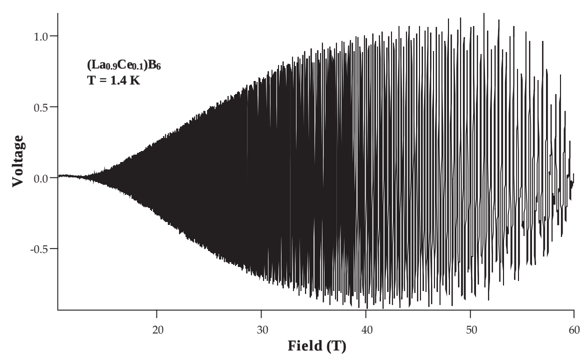

Figure 1. Pulsed field magnetization of $La_{0.9}Ce_{0.1}B_6$, taken at LANL.

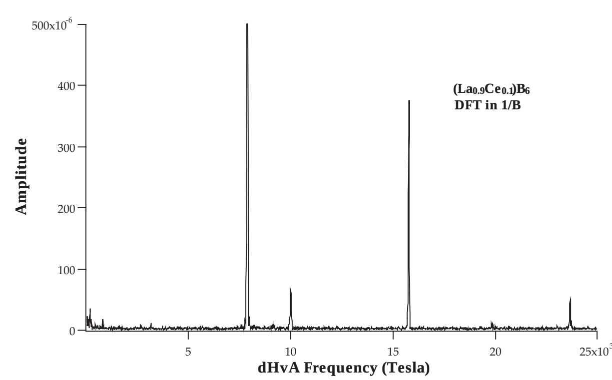

Figure 2. Fourier transform of the data in Figure 1.

the first figure is shown. The fundamental frequency of the α_3 orbit at 7.8 kT has an amplitude of 25 on this scale, but the scale has been reduced to show the $\alpha_{1,2}$ breakdown orbit just above 10 kT and harmonics of the α_3 orbit.

Similar data was obtained for the x = 0.01, 0.05, 0.20, 0.40, and 0.60 samples. Our preliminary analysis of this data shows that the effective mass for the α_3 orbit increases continuously from m* = 0.61 in pure LaB_6 to the pure CeB_6 value of 5.0 at high fields. It is striking to observe that signals of this strength are observed with up to 60% magnetic impurities in a sample, and that the effective mass is increased steadily. It is clear that it may be possible to continue this series all the way to pure CeB_6. In pulsed field measurements low dHvA frequencies with masses greater than one are difficult to observe. In fact, as the Ce concentration was increased, the low frequency hole orbits observed up to 5% Ce were no longer observed in pulsed fields. Therefore, we have made additional measurements in both a 30 T resistive magnet at ^{3}He temperatures and in the 18 T superconducting magnet at mK temperatures in Tallahassee to observe these orbits. The cantilever magnetometer was used in both cases, and a field sweep showing the low frequency oscillations in a 10% sample is shown in Figure 3.

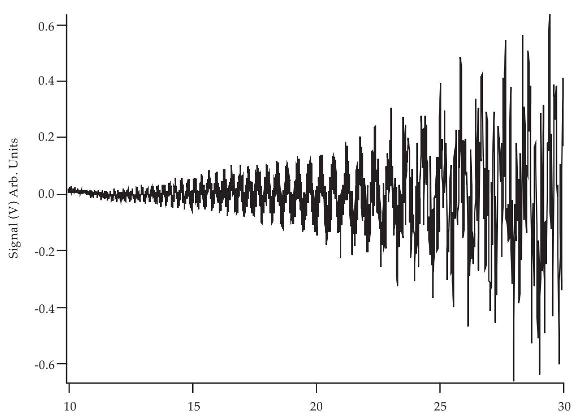

Figure 3. Cantilever torque measurement of dHvA effect in $La_{0.9}Ce_{0.1}B_6$, taken in Tallahassee.

Magnetic Field Dependence of the Effective Mass and de Haas-van Alphen Frequency in CeB_6

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Several years ago we made some preliminary measurements on the dHvA effect on CeB_6. During the past year we extended these measurements from 7 T to 33 T using both the ^{3}He-^{4}He dilution refrigerator and 18 T superconducting magnet system at LSU and the ^{3}He refrigeration system in a 33 T resistive magnet at the NHMFL. Since field modulation measurements were made in each case we were able to make accurate determinations of the dHvA frequency over this entire field range, 7 T to 33 T, and then could calibrate the pulsed field measurements of Haanappel to extend the total range of frequency measurements to 50 T. The result is that after careful analysis, we were able to observe that the dHvA frequency of an orbit on the main piece of Fermi surface (FS) changed in a systematic manner. This variation is shown in Figure 1. In addition, the measured effective masses for this orbit are field dependent, changing from about m* = 30 to m* = 5 between 7 T and 33 T as shown in Figure 2. While

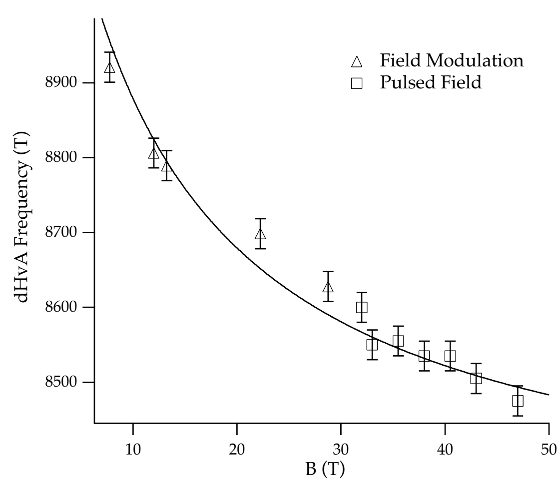

Figure 1. dHvA frequency of the d_3 orbit in CeB_6 as a function of applied field.

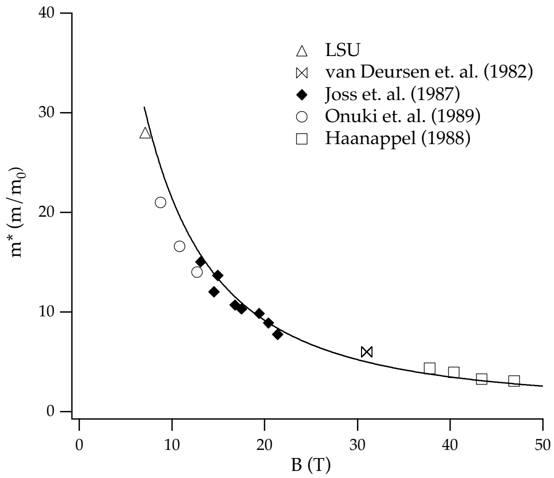

Figure 2. Effective mass of the d_3 orbit in CeB_6 as a function of applied field.

the field dependent mass has been reported before over smaller field ranges, the combination of the field dependent effective mass and field dependent frequency leads to the important conclusion that this piece of FS is a single spin polarized sheet. This fact has important implications for the band structure, and the magnetization in this heavy fermion material. A paper on these results has been submitted to Physical Review, and this work forms the second part of the Ph.D. dissertation by Mr. Donavan Hall.

Quantum Interference in LaB_6

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The quantum interference of electrons in magnetic breakdown networks is a rare phenomenon in metals. The most well known example is that occurring in Mg, often referred to as the Stark quantum interferometer.[1] Since then, the effect has become particularly common in quasi-two-dimensional organic conductors.

Recent magnetization experiments in magnetic fields of up to 60 T have revealed a particularly novel situation in metallic LaB_6, which undergoes magnetic breakdown in fields above ~ 20 T.[2] Since this material is a high conductivity three-dimensional metal, high frequency Shubnikov-de Haas oscillations are difficult to observe in regular transport measurements. Magnetization experiments in pulsed magnetic fields, however, provide an alternative technique whereby oscillations in the resistance, including quantum interference effects, can be observed in the induced eddy currents. One of the observed quantum interference frequencies corresponds to an area in *k*-space exactly equal to that of the Brillouin zone. Since the area of the Brillouin zone is independent of the electronic energy, this frequency also has an effective mass precisely equal to *zero*. Thus the quantum interference oscillations can be observed to very high temperatures. Figures 1a and 1b show examples of Fourier transforms of the induced voltage oscillations measured in LaB_6 at 1.5 K and 70 K respectively. At low temperatures, the de Haas-van Alphen effect dominates, whereas at high temperatures, only the quantum interference frequencies survive.

In spite of the fact that the Brillouin zone frequency has an effective mass of zero, its amplitude is still attenuated at sufficiently high temperatures. This provides valuable information about the effects of phonon scattering in the transport, which is the most likely mechanism that can lead to de-phasing of the oscillations at higher temperatures.

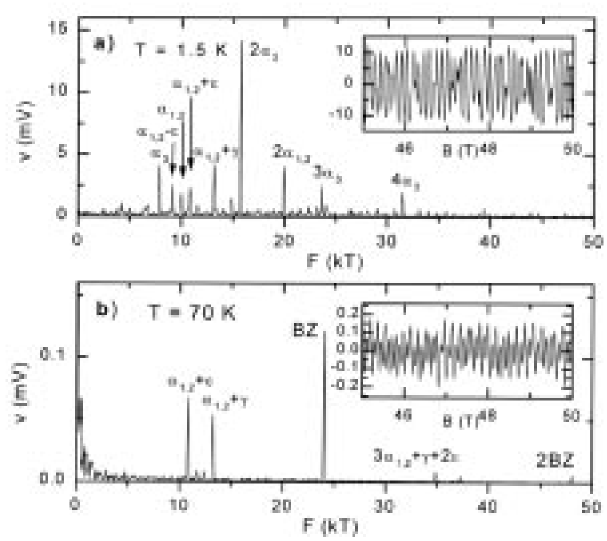

Figure 1. Fourier transforms of the oscillations measured in the detection coils at 1.5 K (a) and 70 K (b). The detected oscillations are shown as insets.

The observation of quantum interference effects in LaB_6 implies the existence of such effects in all three-dimensional metals in which magnetic breakdown occurs. This has far reaching implications for fermiology.

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Quantum Interference of Heavy Fermions

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The recent observation of quantum interference effects in LaB_6 implies the existence of such effects in the heavy fermion compound CeB_6, which has a similar Fermi-surface.[1] The observation of such effects is particularly exciting in CeB_6 because of its strongly correlated ground state.

CeB_6 has been a popular material for study in pulsed magnetic fields[2,3,4] due to the quenching of the effective mass in strong magnetic fields. Nevertheless, no previous experimental investigations have reported quantum interference effects. A number of advances in the experimental capabilities have been made at Los Alamos that enable higher sensitivity measurements than were previously possible. These include the application of pulsed magnetic fields to 60 T while cooling the samples to ^{3}He temperatures. In addition, the magnetization coil detection systems have been miniaturized to overcome effects associated with the inhomogeneity of the magnetic field and sample heating.

Figure 1 shows an example of a Fourier transform of the oscillations measured in CeB_6 at 350 mK, with the oscillations shown in the inset. In many ways, the spectral features are consistent with previous pulsed field investigations.[3,4] Upon expanding the Fourier transform, however, many new features emerge, as shown in Figures 2a to 2d. Many of the combination frequencies that are shown in Figures 2a to 2d cannot be generated by magnetic breakdown alone. The fact that the α_3 frequency (which occurs within a different plane of the Brillouin zone) does not participate in any of the combination frequencies, rules out the effect of magnetic interaction in this material. We can therefore conclude that quantum interference occurs, as has shown to be the case in LaB_6. The most striking feature of the Fourier transform is the zone oscillation frequency in Figure 2d, which

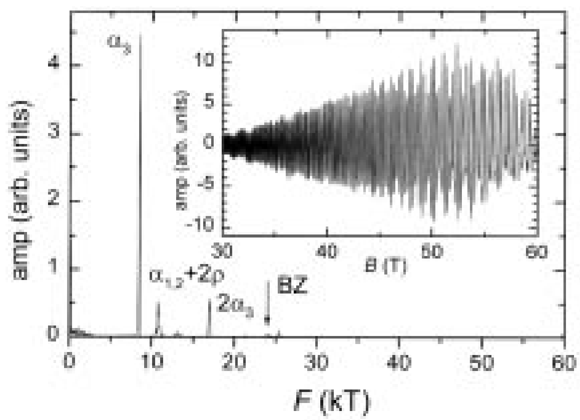

Figure 1. A Fourier transform of the induced voltage in the detection coils from CeB_6 at 350 mK with the oscillations shown in the inset.

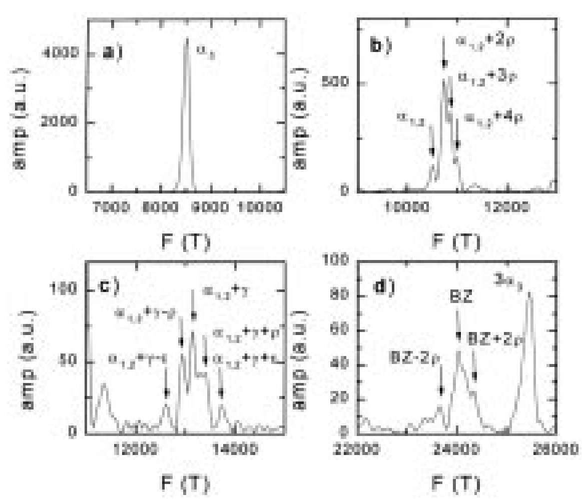

Figure 2. (a)-(d). Expanded views of some of the Fourier transform peaks revealing numerous magnetic breakdown and quantum interference combination frequencies. The Greek symbols refer to the orbits listed in Reference 1.

should have an effective mass of zero irrespective of the heavy fermion mass enhancement.

The results of this study are likely to reveal new information concerning the field dependence of the effective masses for different parts of the Fermi-surface, as well as the overall topological change of the Fermi-surface in strong magnetic fields. Furthermore, the existence of a zone oscillation frequency with an effective mass of zero could in principal enable the potentially exciting possibility of observing quantum oscillatory effects above the coherence temperature.

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Magnetic Impurities in Unconventional Fermi Systems

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One-Dimensional Systems. The experimental study of quantum wires and the quantum Hall effect has led to great activity in the area of one-dimensional Fermi systems. Such systems must be treated within the Luttinger-liquid picture, rather than the more familiar Fermi-liquid (FL) theory, which applies in two and three dimensions. The response of Luttinger liquids to local probes is of particular interest at present. We have studied a generalized Anderson model that includes direct and exchange interactions between localized and delocalized electrons in addition to hybridization between the impurity and the Luttinger liquid. In order to investigate the competition between local-moment formation (and a subsequent Kondo effect) and correlated behaviors arising from strong backward potential scattering,[1] we have mapped the problem onto a classical Coulomb gas which can be analyzed using perturbative renormalization-group methods.

We find[2] that repulsive bulk interactions tend to stabilize local-moment formation by suppressing charge transfer between the impurity and the host. With increasing interactions, the Kondo temperature at first rises, but then falls due to the reduced hybridization. In addition to a magnetic (Kondo) phase dominated by spin-exchange and a non-magnetic phase driven by backward potential scattering, there exists a novel regime that exhibits a Curie susceptibility down to low temperature, but displays the anomalous transport properties of a static impurity. Further work is underway to extend these results into the strong-coupling regime.

Gapless Fermions in 2D and 3D. Another type of unconventional Fermi behavior can arise in spatial dimensions higher than one. Certain anisotropic superconductors, zero-gap semiconductors, and phases of the two-dimensional electron gas in a magnetic field exhibit a quasiparticle density of states that vanishes in a power-law fashion at the Fermi energy. There is now strong evidence that the cuprate high-T_c superconductors belong to this class of materials. Zero-gap HgCdTe is under study as a viable nonmagnetic giant magnetoresistance read-head for high-density storage.

We find that magnetic impurities placed in such systems exhibit a range of remarkable behaviors, both FL and non-FL, that have no counterpart in metallic hosts. Local-moment formation is greatly enhanced by a pseudogap in the density of states, but the conventional Kondo screening of these moments is strongly suppressed. Nonetheless, doping with magnetic impurities may lead to a clear signature of the pseudogap in measurable quantities. Detailed results have been obtained for the thermodynamics.[3] Further work is underway to improve predictions of the transport properties, and to understand a novel class of zero-temperature quantum critical points that arises in these systems.

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High-Field Magnetization Studies of the Heavy Fermion Compound LiV_2O_4

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The transition metal oxide LiV_2O_4 is the first d-metal compound to show magnetic susceptibility, heat capacity, thermal expansion, and NMR properties characteristic of those of the heaviest-mass f-electron heavy fermion materials. This material exhibits a crossover from combined metallic and localized moment behaviors at high temperatures $T > 50$ K to heavy fermion behaviors below about 10 K. In the present work, magnetization M measurements were carried out using DC magnetic fields H up to 20 T and pulsed magnetic fields up to 60 T at temperatures down to 0.5 K in order to further characterize the properties of this unique material. M(H) was found to be linear up to about 20 T, but then to exhibit progressively increasing positive curvature at higher fields. An inflection point in M(H) is found at H = 42 T at 0.5 K to 0.6K, where dM/dH shows a sharp peak. The inflection point stays at about the same H but becomes less pronounced with increasing T. The origin of the anomaly at 42 T is currently unclear, but we note that the characteristic energy associated with 1 Bohr magneton in this field is close to the characteristic (Kondo, spin fluctuation) temperature of about 28 K inferred from other measurements. The magnetization at 60 T and 0.6 K of about 0.7 Bohr magneton/V atom is significantly less than expected for a spin 1/2 with g-factor of 2. The data indicate that antiferromagnetic interactions, as opposed to ferromagnetic interactions, are dominant in LiV_2O_4.

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Magnetotransport of the Heavy Fermion Compound $YbNi_2B_2C$ to 50 T

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From the theoretical point of view, only very models deal with magnetotransport properties in heavy fermion (HF) compounds even in the Fermi liquid regime. In the simplest case of the one-impurity Kondo limit,[1] a negative magnetoresistance is predicted. However, for the case of the Kondo - Lattice,[2] the magnetoresistance is positive for $T > T_K$ and negative for $T < T_K$. Experimentally, it is interesting to note that a positive magnetoresistance is found in only a few HF compounds in the Fermi-liquid regime.

In early measurements of the transverse and longitudinal magnetoresistance to 18T,[3] a large anisotropic magnetoresistance was observed depending on whether the field was applied perpendicular or parallel to the c-axis. We have also found that below ~ 2K, the resistivity in zero field has a quadratic temperature dependence with a magnitude that is consistent with the enhanced specific heat. Both the T^2 coefficient and the impurity contribution the resistivity are field dependent, and quantitatively match theoretical prediction by Chen *et al*.[4]

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Recently we have extended the DC measurements to 50 T (Figure 1) for B perpendicular to the c-axis. Below, 4 K, a shallow local maximum is observed. This maximum in the magnetoresistance moves to lower field as the temperature is decreased. At magnetic fields above 30 T and temperatures below 2 K a slight up-turn to a positive magnetoresistance is observed. This may be due to the intense magnetic field breaking down the strong correlations of the quasiparticles responsible for the HF state.

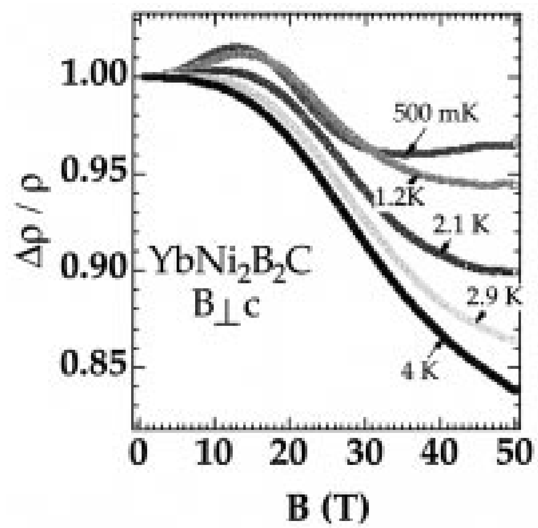

Figure 1. Magnetoresistance of $YbNi_2B_2C$ for B perpendicular to c.

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Magnetotransport in Filled Skutterudites

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Filled skutterudites are cubic materials with the formula RM_4X_{12}, where R = La, Ce, Pr, Nd, Eu, U, or Th, M = Fe, Ru, or Os, and X = P, As, or Sb. Although conventional skutterudites such as $CoSb_3$ are diamagnetic semiconductors, filled skutterudites display a wide variety of ground states, including heavy fermion, ferromagnetic metal and insulator, superconducting, and Kondo insulating.

In filled skutterudites the rare earth or actinide ion sits in an oversized atomic "cage," and appears to undergo large localized vibrations in a shallow potential well. These local modes are effective scatterers of phonons, and have been shown to dramatically reduce the thermal conductivity of the filled materials as compared to their unfilled analogues.[1] Further studies[2] have shown that the specific heat and elastic response of the filled materials are anomalous, and suggest that in the case of La-filled skutterudite antimonides the local mode frequency is about 70 K.

In filled skutterudites, therefore, we have a system in which the rare earth ion (1) undergoes localized, large-amplitude vibrations, (2) has f-orbitals that can strongly hybridize with conduction electron orbitals near the Fermi energy, and (3) can carry a magnetic moment. In such a system, a magnetic field may be able to perturb the local mode frequency of the rare earth ion, and thus interact with the lattice and also (through deformation potential coupling) with the conduction electrons. Large magnetoresistive, magnetostrictive, and magnetothermal effects should be observable.

In our initial set of experiments, we measured the magnetoresistance of $CeFe_4Sb_{12}$ and $EuFe_3CoSb_{12}$ from 2 K to 100 K and in magnetic fields to 18 T. The magnetoresistive behavior of these materials is very different. The magnetoresistance of $CeFe_4Sb_{12}$ is small—positive at low fields and negative at higher fields. The magnetoresistance of $EuFe_3CoSb_{12}$, on the other hand, is very large ($\Delta R/R \approx 20\%$) and qualitatively resembles the magnetoresistance of EuB_6. It is interesting that in EuB_6 the Eu also resides in an oversized atomic "cage."

In the future, we plan to perform magnetoresistance measurements on a wider variety of filled skutterudites, and, if we find the right candidate, perhaps even magnetothermal conductivity experiments.

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Closing the Gap in SmB_6 with Megagauss Fields

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The magnetotransport of SmB_6 has been measured to fields of 145 T at a temperature of ~8 K. The effect of a strong magnetic field on the energy gap of the Kondo insulator SmB_6 (Δ ~ 45 K) has been a great deal of interest for the past couple of years.[1] It has been postulated that a very strong magnetic field will close the gap and the material will re-enter into a metallic state, indicated by positive magnetoresistance. Previous high field measurements to 60 T (0.35 < T < 10 K) have indicated a negative magnetoresistance extending to the highest fields. This investigation has extended the magnetic field by more than a factor of two. The magnetic field was created by use of a single stage flux compression, magnetic field generator.[2] The magnetic field generator, i.e. flux compression, is powered by approximately 4 kg of Detasheet™ C-4 plastic explosives and a 340 kJ capacitor bank. The capacitor bank is used to create the initial current pulse, while the explosives are used to compress the seed flux of the magnet into a smaller volume as well as crowbar the magnet circuit. The seed field risetime (0 T to 20 T) is approximately 50 µs in duration, while a 20 µs risetime occurs between 20 T and 145 T. The experiment terminates shortly after peak field when a jet of vaporized copper contacts the sample area at speeds of approximately 3000 m/s. The magnetotransport was measured by a DC technique that uses six parallel wires (3 for I and 3 for V) to perform a variation of a 4 lead ohmic measurement. The method exploits geometric symmetries which self compensate open loop areas, resulting in a maximum pick-up voltage of 800 mV (in this case). Such levels of pick-up are outstanding considering that the dB/dt induces more than 10 V per square mm at the sample. The sample was cooled by use of a plastic (PVC and G-10) liquid helium cryostat.

A very clean signal was obtained by digitally filtering the time space noise associated with switching transients of the capacitor bank and the magnet crowbar, but the data below 40 T was lost due to these transients. A straight line has been added (dashed), as a guide to the eye, between zero and 40 T. The negative magnetoresistance between 40 and

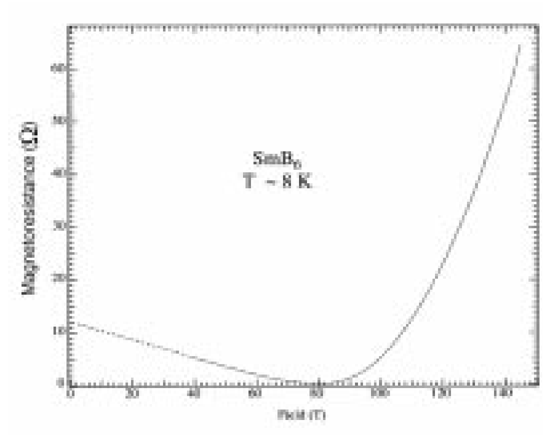

Figure 1. The magnetoresistance of SmB_6 to 145 T. The data below 40 T was lost due to switching transients, a dashed line is inserted as a guide to the eye. The data between 40 and 60 T is consistent with earlier pulsed field measurements in non-destructive millisecond magnets. At ~ 80 T the negative magnetoresistance saturates as the compound regains the properties of a metal. Above 85 T the data suggests the gap is closed as the magnetoresistance becomes positive.

60 T is consistent with earlier measurements. These data indicate the negative magnetoresistance to continue to approximately 80 T at which point the sample resistance is that of a metal (i.e. below 0.5 Ω). Above 85 T metallic behavior is indicated by the positive magnetoresistance. These data are the first indication of closing the gap in SmB_6, however we are cautious considering the incredibly harsh experimental environment so these are considered preliminary data. Further measurements are underway to verify these exciting results. A snubber network (filters) will be added to the capacitor bank on future experiments to help eliminate the switching transient noise.

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B-T Phase Diagram of $CeRh_2Si_2$: Toward an Understanding of the Interplay Between Magnetism and Heavy Fermion Superconductivity

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Heavy-fermion superconductivity is inherently related to magnetism. As such, especially the Cerium based systems $CeCu_2Si_2$,[1] $CeCu_2Ge_2$,[2] $CePd_2Si_2$,[3] $CeRh_2Si_2$,[4] and $CeIn_3$ [5] tend to superconduct in the vicinity of a magnetic ($T = 0$) "quantum critical point," which can be approached, e.g., by application of external pressure. $CeCu_2Si_2$ is intrinsically located in the vicinity of its "quantum critical point": In this system magnetism and superconductivity compete, already at ambient pressure, in the same temperature and magnetic field range.[6] This intriguing interplay between magnetism and superconductivity,[6] however, is not yet understood because of the lack of information about the nature of the magnetism involved. A detailed study of the magnetism has been hampered, so far, by the small size magnetic moment. Yet, the nature of this magnetism might provide a key for understanding superconductivity in these systems.

We studied the isostructural sister compound and local antiferromagnet $CeRh_2Si_2$ (T_N = 37 K). This compound shows heavy-fermion superconductivity under hydrostatic pressure,[4] and, as a new result, also the characteristic magnetic features of $CeCu_2Si_2$, as described in the following.

Utilizing the NHMFL Pulsed Field Facility at Los Alamos, we measured the isothermal magnetization of polycrystalline $CeRh_2Si_2$ up to 50 T. For $T > T_N$ = 37 K we obtain the standard linear contribution of the paramagnet up to high magnetic fields. For temperatures below the antiferromagnetic transition two magnetic phases can be observed: For fields $B <$ 28 T the antiferromagnetic phase (see Figure 1, area

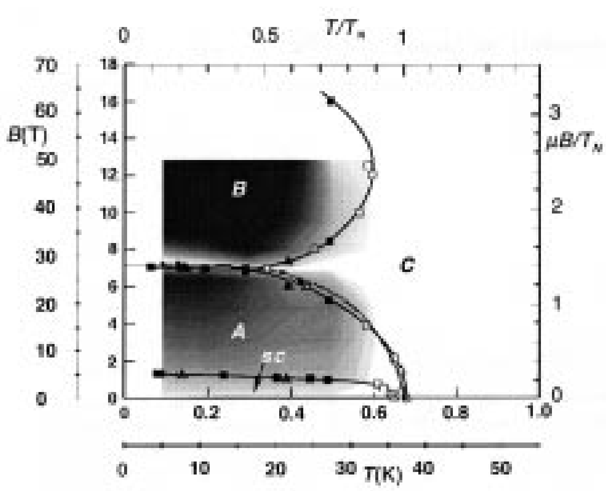

Figure 1. Magnetic phase diagram of $CeRh_2Si_2$ (bullets from oriented powder measurements and shaded areas from polycrystal measurements) and $CeCu_2Si_2$ (squares and triangles from ultrasonic and dilatometric investigations[6]) in scaled units (top and right axis) at ambient pressure. The outer left and lower bottom axis show the actual temperature and field values for $CeRh_2Si_2$, the inner left and upper bottom axis those for $CeCu_2Si_2$. The two magnetic phases are labeled "*A*" and "*B*", the paramagnetic phase "*C.*" The superconducting state (*sc*) only occurs at ambient pressure for $CeCu_2Si_2$. In $CeRh_2Si_2$ the superconductivity sets in under hydrostatic pressure of about 9 kbar.[4]

labeled *A*) displays a decreased magnetization magnitude compared to the paramagnetic state. At $B = 28$ T a first order phase transition into a new high field phase (*B*) takes place that is characterized by its excess magnetization compared to the paramagnetic state (*C*). Based on numerous isothermal high-field magnetization shots, we construct the magnetic phase diagram of $CeRh_2Si_2$ and find a remarkable resemblance to the *B*-*T* phase diagram of $CeCu_2Si_2$ (Reference 6 and Figure 1). We discover, in fact, that the phase diagrams of both systems scale with the size of their magnetic moments μ (Figure 1), leading to the assumption that the magnetism in $CeCu_2Si_2$ is of local 4*f* nature just as in $CeRh_2Si_2$. This finding qualifies to some extent recent speculation about the magnetism in $CeCu_2Si_2$ [6] leading to a more comprehensive understanding of magnetism in the family of Cerium heavy-fermion superconductors.

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Pulsed Field Magnetization Measurements of $CeRu_2Si_2$ to 50 T

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The metamagnetic transition (for magnetic field applied along the c-axis of the tetragonal structure) of the heavy fermion (HF) $CeRu_2Si_2$ has been known for some time.[1] This HF compound presents an enhanced electronic contribution to the specific heat with a Sommerfeld coefficient of ~ 250 mJ/mol K^2, and a Kondo temperature of ~ 25 K. As has been previously pointed out, the large energy separation between the Kondo temperature and the first excited crystal-electric-field level makes this system an ideal compound for studying correlated-electron behavior.

For many years the application of large DC fields (to 23 T) to investigate the thermodynamic and transport properties of this compound has been widely used; however, due to the experimental difficulties, measurements of "good" metals in pulsed magnetic fields have been limited. Here we report for the first time, to the best of our acknowledge, pulsed magnetization measurements in $CeRu_2Si_2$ to 50 T. A clear metamagnetic transition is observed at around 9 T (see Figure 1) consistent with previous DC field data. The magnetization seems to saturate above 35 T. More measurements have to be done to better characterize the saturation of the magnetization and the low frequency oscillations observed.

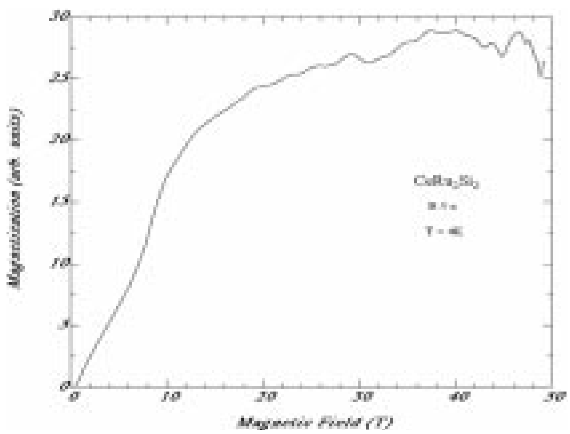

Figure 1. Magnetization of $CeRu_2Si_2$ at 4 K for B // c.

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Pulsed-Field Magnetization Study of $Ce_3Bi_4Pt_3$

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An inductive-type magnetization probe was designed and built to measure the isothermal magnetization of the "Kondo Insulator" $Ce_3Bi_4Pt_3$ in fields to 60 T and at temperatures 1.5 < T < 200 K. The magnetization M of $Ce_3Bi_4Pt_3$ is linear in field at all temperatures, even to 60 T at 1.5 K. Because of this linear variation, M/H vs. T has the same temperature dependence as the low-field susceptibility, which passes through a maximum near 80 K. This maximum in χ is reproduced by a Kramers-Kronig analysis of inelastic neutron scattering data that show the presence of a gap of about 13 meV in the magnetic excitation spectrum at low temperatures. Therefore, one conclusion to be drawn from these high field magnetization results is that 60 T does *not* suppress the spin gap in any significant way. Calibration of the pulsed field data against SQUID measurements to 5 T shows that M(50 T, 4 K) reaches a value only about 10% of that expected for a free Ce^{3+} ion. Clearly, free Ce^{3+} moments do not exist at low temperatures. Instead, the magnitude and field dependence of M agree quantitatively with that expected for a J = 5/2 Kondo impurity having a Kondo temperature T_K = 365 K. This allows us to interpret the origin of the large low-temperature, low-field susceptibility of $Ce_3Bi_4Pt_3$ as arising from field-induced polarization of the Kondo-singlet state. This interpretation does not conflict with the existence of a spin gap, which measures the singlet-triplet excitation energy.

Physics of $YbInCu_4$ and Related Compounds

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As part of an ongoing effort in heavy Fermion physics, we have been studying the first-order valence transition that occurs in $YbInCu_4$. At this transition the magnetic behavior of Yb changes from that of a local moment to a hybridized, mixed-valence configuration, with a corresponding isostructural change in lattice volume. We have mapped the phase diagram of $YbInCu_4$ by studying its response to chemical substitution (Yb, In, and Cu sites), applied physical pressure, and external magnetic fields[1] and have attempted to elucidate the mechanism responsible for this transition by studying related compounds ($YbInNi_4$ and $YbXCu_4$, X=Ag, Au, Cd, Mg, Tl, and Zn).

$YbInNi_4$, which forms in the same crystal structure as $YbInCu_4$, orders ferromagnetically at 3 K and appears to be dominated by crystal field effects rather than the Kondo interaction, as in $YbInCu_4$.[2] In addition to a full complement of zero- and low-field measurements, we have measured the magnetoresistance of $YbInNi_4$ for H < 20 T and 1.5 K < T < 120 K, and have measured the magnetization at 4 K at fields up to 50 T. For 10 K < T < 50 K, the magnetoresistance data can be collapsed on a single curve when plotted as {R(H)-R(0)}/R(0) vs. H/T, consistent with non-Kondo, spin-flip scattering being the relevant scattering mechanism. Below 10 K, ferromagnetic correlations destroy this scaling behavior. Interestingly, despite their strikingly different ground states, $YbInCu_4$ and $YbInNi_4$ have essentially identical magnetization above 40 T. Apparently, once sufficient Zeeman energy is supplied by the external field, the low-energy details of both systems are lost and a magnetization consistent with Yb's full multiplet is recovered.

Many $YbXCu_4$ compounds, where X is a semimetal or late transition metal, form in the cubic C15b crystal structure and display a variety of ground states, including magnetic order and mixed valence, in addition to the isostructural valence transition observed for X=In. We have performed a systematic study of the thermodynamic and transport properties of $YbXCu_4$ for X=Au, Zn, Ag, Cd, In, Mg, and Tl, including high-field magnetization and magnetoresistance. These materials have Kondo temperatures that range from less than 10 K to greater than 900 K. The corresponding saturated magnetization at 50 T varies from 3.9 μ_B to 0.14 μ_B. We are examining whether the conventional single-impurity-theory description can provide an adequate explanation of both the bulk magnetization and the temperature dependence of the f-level occupation number, as deduced from L_{III} x-ray absorption. The extent to which carrier density plays a role in determining the physical properties of these materials is also being studied. Through this effort, we hope to better understand not only the valence transition in $YbInCu_4$ but also the more general aspects of correlated electron phenomena.

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Magnetization of UBe_{13} to 60 T

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One of the more remarkable aspects of the heavy fermion superconductor UBe_{13} is the linearity of its isothermal magnetization in high magnetic fields. Early characterizations of the magnetization specify deviations from linearity in terms of a few percent at some large field. We have measured the magnetization of UBe_{13} in pulse fields to 60 T at 4.0 K in an effort to extract more quantitative information on the shape of M(H) and to search for possible metamagnetic transitions such as have been observed in, for example, UPt_3.[1]

We studied a powdered sample of UBe_{13} (grain sizes from 125 to 250 μm) to minimize eddy current heating. Measurements of M(H) using two different inductive pickup probes and two different pulse magnets yielded consistent results at 4.0 K. The magnetization at 50 T, about 1 μ_B/U-atom, is still well below the high temperature moment of 3.4 μ_B/U-atom determined from a SQUID magnetometer on a solid chunk of the same sample. M(H) at 50 T is suppressed by about 14% below linearity with no sign of a metamagnetic transition to 60 T.

We fit the data to extract the nonlinear contribution to the susceptibility in a manner described elsewhere.[2] Even to these high fields the data is well fit with a single nonlinear term. We found χ_3=-0.05 emu/T^3 in reasonable agreement with earlier results[2] after correction for the temperature dependence of χ_3.[3] This agreement with earlier results combined with the agreement between the M(H) data in increasing and decreasing fields indicates that self heating is not a problem with our sample at this temperature. We plan to extend these measurements to higher fields and lower temperatures to extract the saturation magnetization of UBe_{13} and to search for possible metamagnetic transitions.

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Upper Critical Magnetic Field of Boron Doped UBe_{13}

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As Boron is doped onto the Beryllium sites of UBe_{13} the discontinuity in the specific heat at T_c doubles in size (at a Boron concentration of about x=.002 in $U(Be_{1-x}Bx)_{13}$) while T_c itself does not change within experimental resolution.[1] We measured the upper critical magnetic field $H_{c2}(T)$ of a polycrystal sample of in $U(Be_{1-x}Bx)_{13}$ with

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x=0.00223 to further investigate the effect of Boron doping on the superconducting state.

We used a standard four terminal resistance technique and measured $H_{c2}(T)$ (defined as the midpoint of either an isothermal or iso-field resistance transition) from about 20 mK to T_c = 0.89 K. We find an initial slope of about –18 T/K, low temperature saturation toward $H_{c2}(0)$ = 9.0 T, and an overall shape similar to that of UBe_{13} doped with other impurities.[2]

The initial slope we found is less than half that observed for pure UBe_{13},[3] which is surprising in light of the fact that the electronic coefficient of the specific heat (and hence the density of states) appears to be unaffected by Boron doping for this concentration and the fact that the enhanced jump in the specific heat suggests an enhanced coupling.[1] Our resistance transitions, however, show the effects of a small impurity phase that complicates the interpretation of our results at this time. Further measurements are planned on samples with a wide range of Boron concentrations.

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High Field Scaling Experiments in nFl Systems

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We visited the NHMFL twice to measure specific heat and once to measure magnetization. The latter measurement must be repeated—it was too noisy. For the specific heat, we succeeded in measuring a standard sample (gold) to check to see if the rig gave good data up to 32.9 T. We then measured five research samples. Most interesting were $U_{1.2}La_{0.8}Zn_{17}$, which is a non-Fermi liquid system, and $CePd_2Si_2$, which is an antiferromagnet at 10 K. On the Zn sample, we determined the scaling exponent beta to be 0.7 in fields of 13, 17, 21, 25, 29, and 33 T. For the $CePd_2Si_2$, we measured specific heat in 29 T and determined that $T_{(Neel)}$ is not suppressed appreciably—the transition in the specific heat is broadened, but stays more or less pinned to the same temperature.

Successive Metamagnetic Transitions and Magnetoresistance in the Low-Carrier-Density Strongly Correlated Electron System CeP

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Successive metamagnetic transitions in CeP are studied through magnetoresistance measurements up to 31.5 T over a wide temperature range between 0.7 K and 44.1 K (Figure 1). The resulting magnetic phase diagram is considerably different from that of a previous report.[1] In the present case, the transition fields are not equidistant in inverse magnetic fields, which does not support the previous idea that those transitions are triggered by crossings of the up- and down-spin Landau levels in a particular electronic energy band. The present phase diagram reveals its close similarity to that of CeSb and its general features are satisfactorily explained with a simple thermodynamic model.

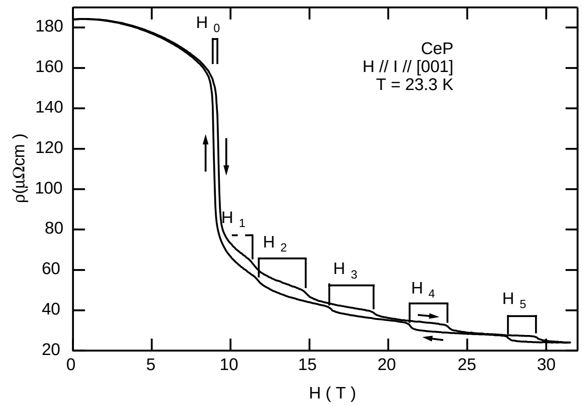

Figure 1. Resistivity of CeP at T = 23.3 K as a function of the magnetic field. The magnetic field and the electric current were parallel and applied along the [001] direction.

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